

Ozone differences between near-coastal and offshore sites in New England: Role of meteorology

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[1] Time series from two ozone monitoring stations are evaluated, one on an island several km off the New England coast, the other several km inland in New Hampshire. In the summer of 2002, during the New England Air Quality Study 2002 (NEAQS-2002), ozone measurements at the island station, Appledore Island (ADI), were consistently higher than at the inland station, Thompson Farm (TF). We hypothesized that the differences in ozone concentrations were due to transport differences driven by mesoscale meteorology, since neither site was in a source region. We found that the Appalachian Trough, coastal cold fronts and coastal stationary fronts at times caused TF to have westerly component flow while ADI had southerly component flow. In these situations, the southwesterly flow along the New England coast brought ozone and precursors to ADI from metropolitan areas to the southwest (e.g., Boston). Conversely, the air transported to TF from the west was contaminated by fewer upstream sources, and therefore the ozone was lower at TF. The sea breeze was also a factor, which tended to have the contrasting effect of nearly equalizing the ozone concentrations at the two sites by transporting ozone-rich air already impacting ADI inland to TF. Enhanced measurements from the NEAQS-2002 study were used in the analysis, including radar wind profilers, Doppler and ozone profiling lidars, and radiosondes launched from a ship. We also assessed model performance for two models, WRF/Chem and MM5/Chem, for four key days.

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1. Introduction

[2] Differences in meteorological processes on local scales can lead to systematic differences in pollutant concentrations between sites only a few tens of km apart. One area where such differences in meteorology can occur is near seacoasts. In this study we investigate differences in ozone concentrations during summer 2002 that often exceeded 20 ppbv or more between two seacoast sites. These sites are separated by 30 km, one an island station 10 km off the coast of New Hampshire, the other a station 20 km inland. Extensive atmospheric chemistry measurements were taken at these sites by the University of New Hampshire (UNH), as part of the New England Air Quality Study (NEAQS-2002,

<http://www.esrl.noaa.gov/csd/NEAQS/>) and the Atmospheric Investigation, Regional Modeling, Analysis and Prediction program (AIRMAP [Mao and Talbot, 2004a]). These projects were designed to increase understanding of the atmospheric processes that control the production and distribution of air pollution in the New England region of the United States (US). AIRMAP is an ongoing project at UNH and NEAQS-2002 was a summertime study that involved many agencies and included additional instrumentation in the region during July and August of 2002 [Angevine et al., 2004; Bates et al., 2005; Brown et al., 2004; Dibb et al., 2004; de Gouw et al., 2003].

[3] The two sites we investigate are Appledore Island, ME (ADI) and Thompson Farm, NH (TF, Figure 1). With ADI experiencing marine conditions, and TF experiencing continental or modified marine conditions, the investigation of the differences in ozone between these two sites allows us to evaluate regional transport of ozone and its precursors over both land and water. Over the course of the summer of 2002, the differences in ozone between the two sites often exceeded 20 ppbv, and occasionally the differences were as much as 50 ppbv or more, with ADI consistently having the higher ozone concentrations.

[4] Griffin et al. [2004] have established through a study of three ozone formation metrics (instantaneous and net

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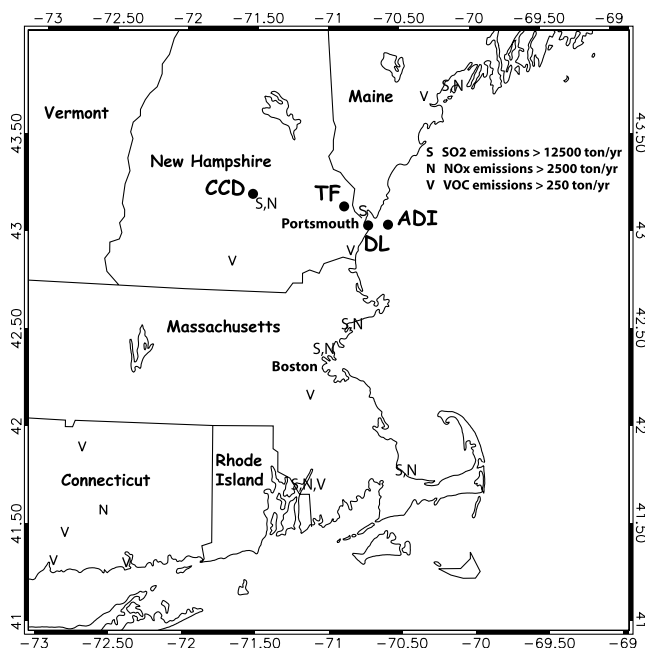


Figure 1. Map of study area. Locations of Thompson Farm (TF), Appledore Island (ADI), the Doppler lidar at Rye Harbor (DL), and the Concord, NH, wind profiler (CCD) sites are marked with a dot. For general reference, significant sources of SO₂, NO_x, and VOCs are indicated by S, N, or V, respectively.

ozone production rates, instantaneous and average ozone production efficiencies, and hydrocarbon and carbon monoxide reactivity) that high ozone concentrations occurring during August 2002 at TF were most likely the result of the transport of ozone-rich air rather than local production. This is based on their findings that the abundance of biogenic alkenes point to few local anthropogenic emissions sources, and the instantaneous and net ozone production rates at TF were relatively low compared to other locations within the US and cannot account for the large ozone peaks that occurred at TF in summer 2002.

[5] If transport processes control the ozone observed at TF, and ADI is an island with no local sources only 30 km southeast of TF, it is likely that transport controls ozone dynamics at ADI also. This leads to our hypothesis that differences in ozone transport play a large role in the differences in ozone between the two sites, in conjunction with other factors such as nighttime differences in deposition and titration. Since the sites are ~30 km apart, these differences in transport are likely to be associated with regional-scale or mesoscale meteorology, including the differences in transport over water versus over land.

[6] Synoptic-scale meteorological processes, events that last more than a day and have a horizontal spatial scale ranging from several hundred km to several thousand km [American Meteorological Society, 2000], can create the setting for high pollution events through large-scale subsidence, leading to an increase in sunshine, weak winds and reduction in precipitation (all associated with high ozone days in major metropolitan areas, but not rural New England). The synoptic-scale meteorology also defines the long-range transport, which is the key for New England air

quality because of the high ozone concentrations that are measured there in spite of the lack of local sources. However, smaller-scale mesoscale-meteorological processes, particularly the meso-beta and meso-gamma scales, often determine the final outcome for pollution levels in one area versus another, or one day versus another. (Meso-beta-scale events last less than a day and have a horizontal spatial scale from 20 to 200 km, while the meso-gamma scale includes smaller events that last less than a day and have a horizontal extent ranging from 2 to 20 km [Orlanski, 1975].) A well-documented example of a mesoscale event, and its effect on air quality, is the sea breeze [e.g., Angevine et al., 2004; Banta et al., 2005; Darby, 2005; Flocas et al., 2003; Gaza, 1998; Lu and Turco, 1994; Lyons and Olsson, 1973].

[7] Gaza [1998] gives an overview of the importance of mesoscale meteorological phenomena in New England air quality, including lee-side troughs, fronts, and sea breezes. It was shown that over five summers in New York state, a majority of high ozone days had a trough or surface front in the region of the high ozone. The frontal boundary or trough served to “focus” the ozone concentrations near the trough axis through converging winds, with the highest ozone ahead of the trough. Our analysis will show that a front or trough located near the New England coast played an important role on the horizontal distribution of ozone, influencing the amount of ozone measured at TF and ADI, consistent with Gaza’s [1998] analysis of New York stations. The important effects the Appalachian lee-side trough [Weisman, 1990] has on air quality in New England has been shown by Seaman and Michelson [2000] and Mao and Talbot [2004a].

[8] Gaza [1998] also makes the important point that air quality models will not give accurate ozone forecasts if the mesoscale meteorology is not well represented in the model initialization stage or adequately forecast. We evaluate how two different models, the Weather Research and Forecasting/Chemistry model (WRF/Chem [Grell et al., 2005]) and the Mesoscale Meteorological Model/Chemistry, version 5 (MM5/Chem [Grell et al., 2004]), performed during four days of August 2002.

[9] Mao and Talbot [2004a] assessed transport processes to inland and coastal sites using a mesoscale numerical model with chemistry. We build on their concepts regarding ozone differences between ADI and inland stations, but with more emphasis on detailed observational analyses, including enhanced observations from the NEAQS-2002 deployments. For instance, through radar wind profiler measurements we evaluate measured mixing heights and infer mixing vigor, rather than using modeled mixing heights. Also, we will use the NOAA wind profiler trajectory tool [White et al., 2006] to create surface trajectories and trajectories aloft to assess transport at more than one level. These back trajectories were based on measurements, rather than model output.

[10] In section 2 of the paper we describe the TF and ADI sites and give an overview of the ozone measurements from July and August 2002. In section 3 we present a detailed analysis of four days in August, all with differing mesoscale meteorological settings. In section 4 we present back trajectories derived from a network of radar wind profilers and buoys that help to elucidate the transport patterns

associated with the differences in ozone between TF and ADI. We give an overview of how mesoscale models handled our four key days in section 5. Section 6 contains a summary.

2. Site Description and Overview of Summer 2002

[11] TF is located south of Durham, NH, ~20 km inland (70.95W, 43.11N), at 24 m above sea level (Figure 1). Air quality instruments were mounted on a 40-foot tower, on a site with rolling hills surrounded by mixed forest. Measurements at this site included those of both continental and marine air masses. Instrumentation and measurements at the site are described in detail by *Mao and Talbot* [2004a, 2004b, 2004c], *Griffin et al.* [2004], and *Talbot et al.* [2005], who also briefly describe the TF site.

[12] The ADI site is part of the Shoals Marine Laboratory on Appledore Island, ME (70.62W, 42.97E), ~10 km from the New Hampshire coast, at sea level, and ~30 km southeast of TF. The island is part of a small group of islands, called the Isles of Shoals. ADI is small, ~0.6 km wide (east-west) at its widest point and ~0.94 km long (north-south). Measurements are taken only during warmer months when the island is inhabited. During NEAQS-2002, a radar wind profiler, a meteorological surface station, and chemistry and aerosol sensors were operated (deployed by the University of New Hampshire and the NOAA's Earth System Research Laboratory (NOAA/ESRL)). The instrumentation and methods for measuring ozone and carbon monoxide at ADI were identical to those of TF. The island's location allows for measurements of pollution plumes advected over water from source regions to the south and southwest of ADI, as well as continental and marine air masses transported from other directions.

[13] Ozone measurements from ADI and TF from 1 July to 31 August 2002 are shown in Figure 2a. It is immediately apparent that ozone at ADI (red trace) was generally higher than at TF (black trace). The differences in the daytime measurements could be quite large between the two sites, with ADI ozone exceeding TF ozone by 20 ppbv or more at times (Figure 2b). At night, TF's ozone tended to drop several ppbv lower than ADI's. These characteristics are further exemplified by the scatterplots of daily ozone maxima and minima shown in Figures 2c and 2d, respectively, for the same time period shown in Figures 2a and 2b.

[14] Figure 2b shows ADI ozone minus TF ozone. On average, ozone was 11.6 ppbv higher at ADI for this period, with a standard deviation of 13.6 ppbv. When divided into daytime and nighttime hours, the mean difference between the two sites was 9.4 ppbv with a standard deviation of 13.5 ppbv during the day, and 14.7 ppbv with a standard deviation of 13.2 ppbv at night.

[15] The scatterplot for ozone maxima for the two sites (Figure 2c) indicates that it was unusual for the daily maximum ozone at TF to exceed that of ADI, and then only by a small amount. The Pearson correlation coefficient for ozone maxima between the two sites was $r = 0.88$. Although there was a high correlation between the two sites, the linear fit between the two sites fell below the one-to-one line because the ozone at ADI was consistently higher than at TF. Days when the maximum ozone at ADI was >80 ppbv

fell into two groups of days: days when both sites had similar maxima (blue stars) and days when ADI had a much higher maximum than TF (red stars). Most of the mid-August days when ADI and TF had similar maxima >80 ppbv were described in *Angevine et al.* [2004], who showed that three of these days had a sea breeze that advected ozone-rich air inland, leading to high ozone at both sites. It is the second group of days, when ozone maxima at ADI were much higher than at TF, that is of interest in the present analysis.

[16] The ozone minima at both sites had a much weaker Pearson correlation coefficient, $r = 0.56$ (Figure 2d). *Talbot et al.* [2005] attribute nighttime ozone loss at TF in summer to deposition (~11 ppbv per night) and titration by NO (~20 ppbv per night). The titration of ozone by NO is related to vehicular traffic and other surface combustion sources, and deposition of ozone over water is far less than that over land. Therefore these ozone removal mechanisms, which play an important role at TF at night, do not exert as much influence at the small island site of ADI, explaining the differences in the ozone minima between the two sites. The fact that ADI is more likely to begin the day with more ozone may also contribute to higher daytime ozone values.

[17] The days with the most extreme ozone differences were 2–4 July and 13 and 17 August 2002, with ADI ozone exceeding TF ozone by 68 ppbv or more (Figure 2b). These extreme differences typically occurred after the ozone had peaked at ADI, and the ozone at TF had started its rapid decline near the time of sunset, further enhancing the already large daytime difference in ozone between the two sites. Although part of the AIRMAP data set, none of these days occurred during the more intensive measurements of the NEAQS-2002 field program.

[18] Analysis of synoptic weather maps for these extreme days (not shown) revealed an Appalachian lee-side trough for all of these days. This type of trough, which has been shown to enhance transport of pollution northeastward, along the U.S. east coast [*Seaman and Michelson*, 2000; *Mao and Talbot*, 2004a], was also present on 15, 18, and 23 July 2002 when the ozone differences between the two sites exceeded 50 ppbv.

[19] Much smaller differences between ozone concentrations at the two sites occurred in the days immediately following the Appalachian trough, due to cold front passages, which reduced ozone regionally by bringing in an air mass from lower-ozone areas, and by shutting down the southwesterly transport along the coast. This phenomenon has been described in previous air quality work [e.g., *Cooper et al.*, 2001; *Moody et al.*, 1996; *Mao and Talbot*, 2004a].

[20] In this paper, we focus on 3–6 August, days which fall into the NEAQS-2002 field program. The ozone differences between ADI and TF on these days were smaller than on the extreme days, 40 ppbv or more at times. *Mao and Talbot* [2004a] previously investigated differences in ozone between ADI, TF, and three other sites, two on the coast and one further inland than TF, during summer 2001. Through modeling and data analysis, *Mao and Talbot* [2004a] suggest that reasons for greater ozone at ADI include more diverse upwind sources for ADI, lower mixing heights over ADI leading to less vertical dilution than at TF, and transport of ozone by the sea breeze. They also suggest

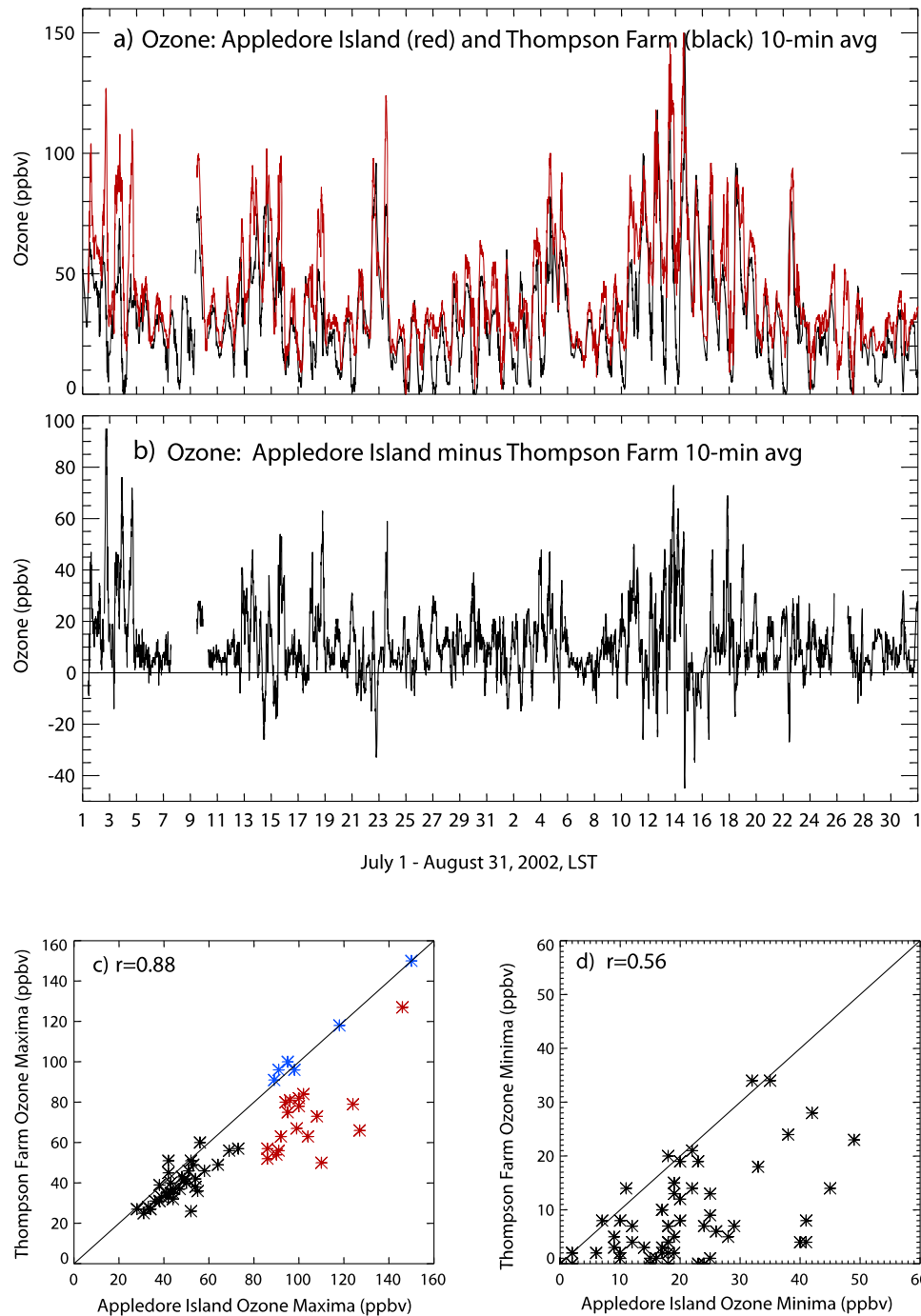


Figure 2. (a) Time series of 10-min averages of 1-min ozone data from ADI (red) and TF (black). (b) Time series of ADI ozone minus TF ozone. (c) Scatterplot of ozone maxima for each day from 1 July to 31 August 2002, TF versus ADI. Blue stars represent maxima that occurred on 22 July and 11–12, 14–15, and 18 August. Red stars include many days between 1 July and 22 August when the difference between ozone maxima at the two sites was large. (d) Scatterplot of ozone minima for each day from 1 July to 31 August 2002, TF versus ADI. Ozone data were downloaded from <http://airmap.unh.edu/DownloadData>.

that during the afternoon, when there is larger-scale southwesterly flow converging with onshore sea breeze flow, pollutants from metropolitan areas such as Boston can become elevated at the convergence zone, as demonstrated near the Texas seacoast by Banta *et al.* [2005], where they can then be more quickly advected toward ADI. We use the modeling results of Mao and Talbot [2004a] to guide our

detailed observational investigation of the differences in ozone between ADI and TF.

3. Measurements 3–6 August 2002

3.1. Overview of 3–6 August

[21] In this section, we present time series of chemical and meteorological variables for 3–6 August 2002, to take a

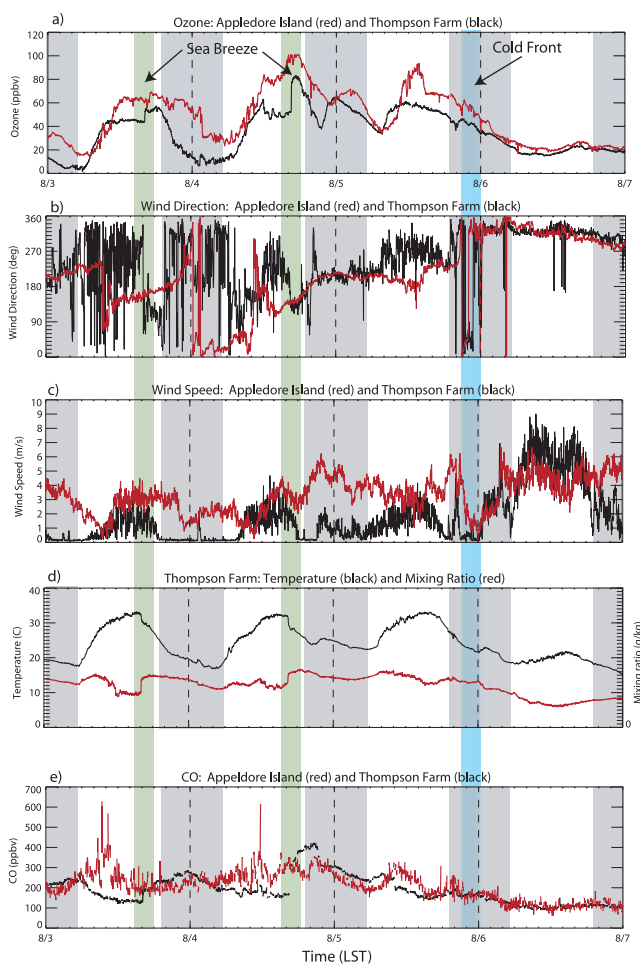


Figure 3. Time series of various quantities for 3–6 August 2002. (a) Ozone (ppbv) at ADI (red) and TF (black). (b) Wind direction (degrees from north) at ADI (red) and TF (black). (c) Wind speed (m s^{-1}) at ADI (red) and TF (black). (d) TF water vapor mixing ratio (g kg^{-1} , red) and TF temperature ($^{\circ}\text{C}$, black). (e) CO (ppbv) at ADI (red) and TF (black). Nighttime hours are shaded light gray. Green shading indicates the transition time surrounding the onset of the sea breeze at TF. Blue shading indicates the transition surrounding the passage of a synoptic-scale cold front. Meteorological data were downloaded from <http://airmap.unh.edu/DownloadData>.

closer look at the ozone behavior at both ADI and TF on these days and to determine reasons for the higher ozone at ADI. We use synoptic-scale observations to provide a larger-scale setting for the interpretation of the local meteorological data, with an emphasis on transport pathways for each site. We also investigate the effect of the sea breeze on measurements at TF, as well as mixing height differences between TF and ADI. Figure 3 shows ozone, wind direction, wind speed, and carbon monoxide (CO) from both ADI (red) and TF (black) for 3–6 August 2002. Figure 3 also shows temperature (black) and mixing ratio (red) from TF.

3.2. Winds, Ozone, and CO for Each Day

[22] Both 3 and 4 August were sea breeze days at TF as indicated by a wind shift to onshore (southeasterly) flow

perpendicular to the coast (Figure 3b) and a coincident drop in temperature and rise in water vapor mixing ratio (Figure 3d), standard meteorological characteristics indicating sea breeze onset [Atkinson, 1981; Banta *et al.*, 1993]. Accompanying the sea breeze onset at TF was a sharp increase in both ozone and CO on both days (Figures 3a and 3e). The increase in CO indicated a change in air mass to one that originated in a metropolitan area, whereas the increase in water vapor mixing ratio and shift to southeast winds indicated that the air mass was advecting inland from the ocean. Combined, these meteorological and air chemistry data show that urban pollution was transported over water, and then brought in by the sea breeze, as suggested by Angevine *et al.* [2004].

[23] Meteorological data also provided evidence for a sea breeze onset at ADI on both 3 and 4 August at ~ 1130 LST, with both days showing a slight drop in temperature (not shown) coincident with the onset of steady southeasterly flow (Figure 3b). A forward differencing analysis of the TF and ADI ozone time series was performed in which the difference between each measurement at time t and the measurement at time $t + 10$ min was plotted as a function of time. This analysis (not shown) indicated that the sea breeze on 4 August caused the largest jump in ozone over a 10-min period at TF in the entire two months analyzed. This analysis also indicated that the sea breeze onset had no impact on ozone at ADI on 3 or 4 August.

[24] A map showing synoptic-scale data for 1300 LST 3 August is shown in Figure 4a. 1300 LST was chosen to portray afternoon conditions during the time of the largest daytime ozone difference between TF and ADI, and before the sea breeze onset at TF. The primary features in Figure 4a were a stationary front hugging the New England coastline and a broad area of high pressure to the west, behind the front. ADI experienced prefrontal southerly component flow, providing transport of pollutants from sources to the south and southwest of ADI. TF, behind the stationary front, had westerly flow prior to the sea breeze onset (Figure 3b), indicating transport from a region with far fewer pollution sources. Thus both sites were in different air masses and subjected to different transport pathways, leading to higher ozone concentrations at ADI. Figure 5a summarizes the situation for the day, showing eastward transport of pollution from Boston (KBOS) in the morning (small light gray arrow), priming the pollution for transport to ADI, a sea breeze at both KBOS and ADI at 1300 LST and pre-sea-breeze westerly flow at TF (small dark gray arrow), and flow aloft from the northwest (large light gray arrow).

[25] The synoptic map for 4 August (Figure 4b) indicated that the stationary front from the previous day had dissipated, and the dominating feature near the east coast was a weak surface trough aligned along the Appalachian Mountains, the Appalachian trough. The overall weak synoptic-scale forcing allowed the sea breeze to form again on 4 August. Before the sea breeze onset at TF, ADI experienced southeasterly sea breeze flow, whereas TF experienced westerly component flow, similar to the situation on 3 August. What was different from 3 August was that the early morning pollution export from Boston was to the southwest, away from TF and ADI, and winds aloft were southerly rather than northwesterly (Figure 5b). Combined, these factors indicated possible enhanced pollutant transport

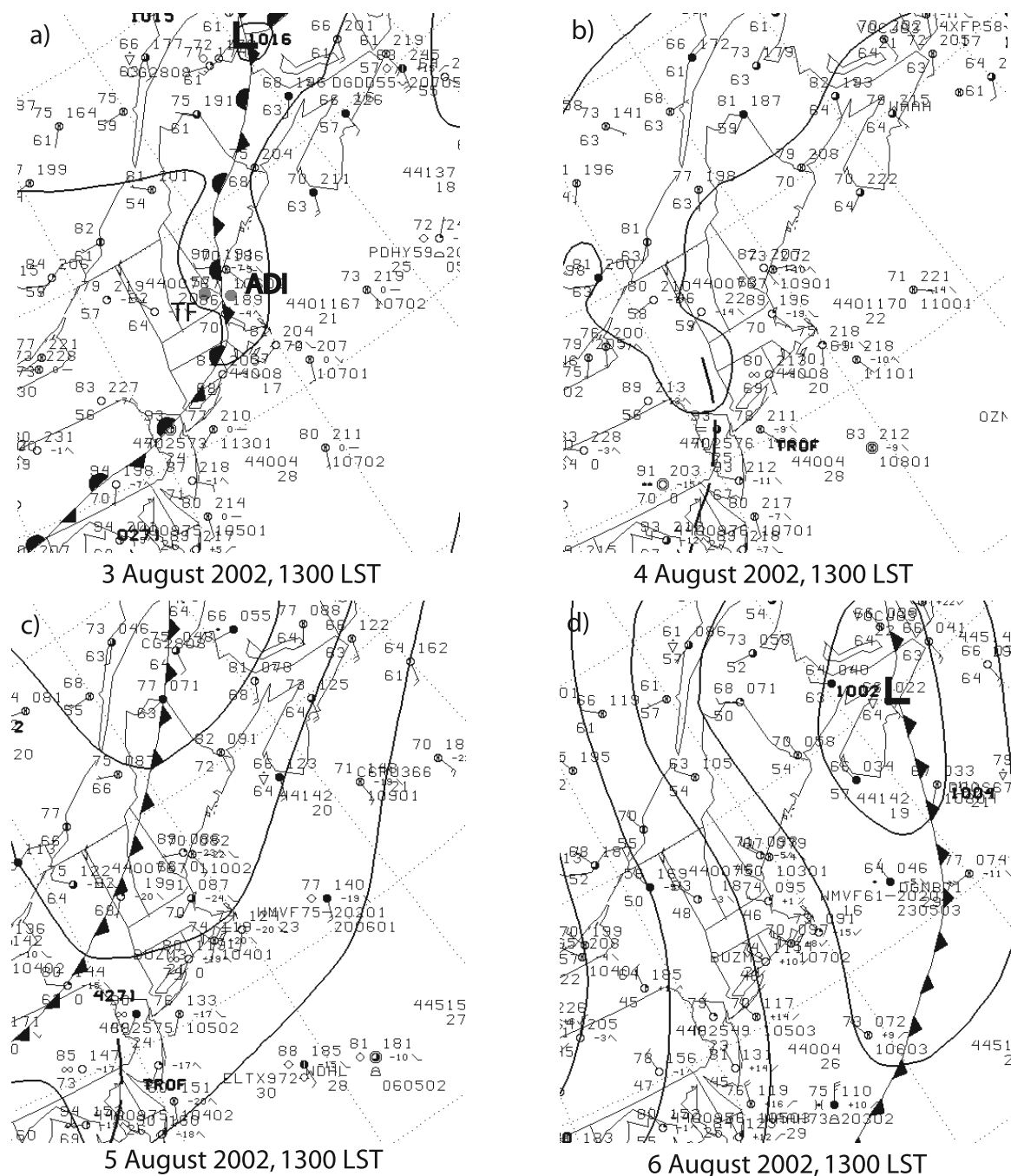


Figure 4. Surface synoptic maps for 1300 LST 3–6 August 2002. (a) 3 August 2002. Locations of ADI and TF are given by gray dots. (b) 4 August 2002. (c) 5 August 2002. (d) 6 August 2002. All symbols are standard synoptic weather analysis symbols. Maps were acquired from the NOAA/National Climatic Data Center.

aloft. Ozone profiles from the Ozone Profiling Atmospheric Lidar (OPAL) system indicated that there was relatively high ozone above the surface during daytime hours of 4 August (Figure 6). Figure 6b shows ozone time series from the deck of the R/V *Ronald H. Brown* (which is essentially the same as at ADI) and from OPAL profiles averaged between 300 and 500 m ASL. In the hours before noon it was apparent that there was a reservoir of ozone aloft, available for downward transport.

[26] 5 and 6 August were not sea breeze days (Figure 3). The surface trough that started to form on 4 August was fully developed on 5 August (Figure 4c), and thus this whole day was a pre-cold-front/Appalachian trough day with large-scale southwest flow along the eastern seaboard of the US, transporting pollutants to the New England sea coast (Figure 4c). The wind direction time series (Figure 3b) indicated that throughout much of the day ADI had predominantly southerly flow, whereas TF had predominantly westerly flow. The differences in midday ozone and CO

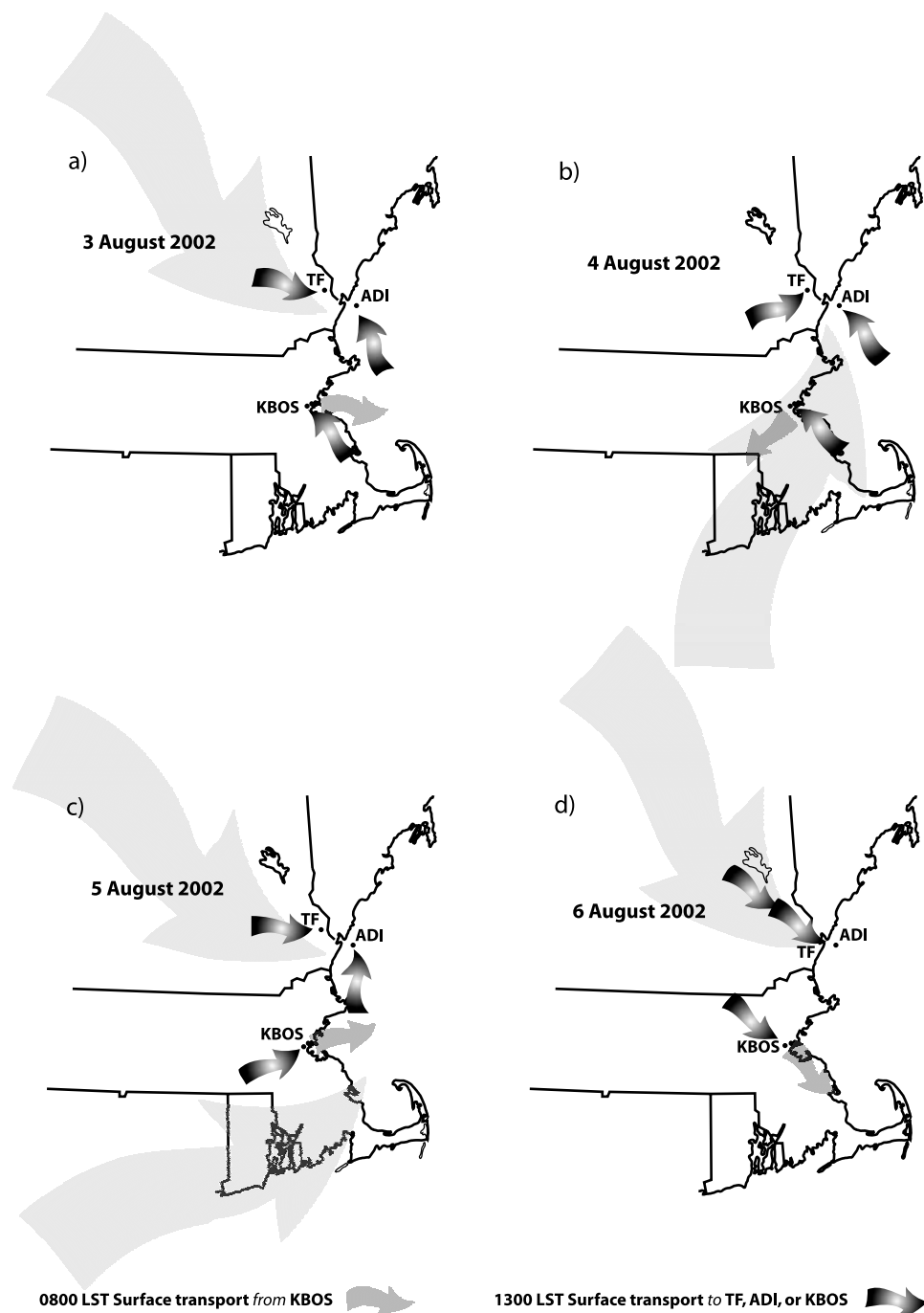


Figure 5. Overview of surface and 925-mbar winds for 3–6 August 2002. Small gray arrow indicates the direction of pollution transport from KBOS based on the 0800 LST National Weather Service (NWS) observations. The small black arrows with white centers indicate the transport toward KBOS, TF, or ADI based on the wind direction at 1300 LST. The large light gray arrow indicates transport aloft, at 925 mbar, based on NWS soundings at 1900 LST from Chatham, MA, Portland, ME, Albany, NY, and Long Island, NY.

between TF and ADI were a result of these differing wind directions, indicating that each station was on opposing sides of the trough axis. Figure 5c summarizes this. Without the sea breeze transport of additional ozone to TF, the TF ozone time series did not have much variation after the morning rapid rise, and was 40 ppbv lower than ADI in the afternoon.

[27] Late on 5 August the synoptic-scale cold front seen in Figure 4c swept through the region from the north-northwest, reducing ozone and CO at both sites because of the lack of significant pollution sources to the north-northwest of these sites (Figure 1). Synoptic conditions on 6 August, when moderate post-cold-front northwest winds dominated, provide a contrast to the other three days when

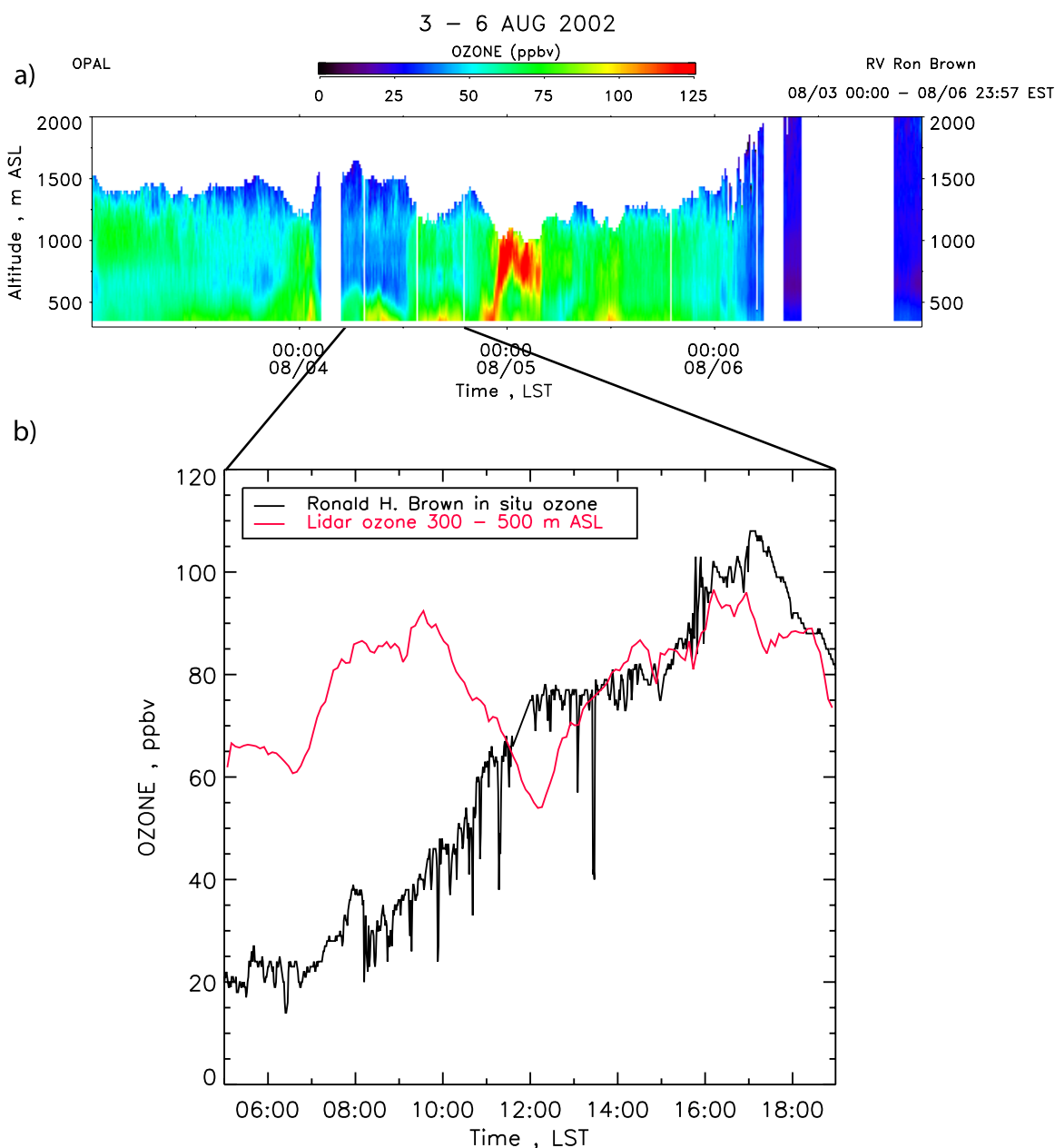


Figure 6. (a) Time-height display of ozone profiles from the Ozone Profiling Atmospheric Lidar (OPAL) system from 0000 LST 3 August 2002 to 0000 LST 7 August 2002. Color scale indicates ozone concentrations in ppbv. (b) Ozone time series derived from ozone profiles averaged between 300 m and 500 m ASL (red) and in situ ozone measurements from the deck of the R/V *Ronald H. Brown* (black) from 0500 to 1900 LST on 4 August 2002. Both ozone measurement systems were deployed on the R/V *Ronald H. Brown* by NOAA/ESRL.

mesoscale features, i.e., the sea breeze or the Appalachian trough, dominated.

[28] Another interesting feature in the chemistry time series occurred about 2 hours before midnight on 5 August (Figure 3). Ozone rose sharply and CO declined steeply at TF. Since this occurred at night when photochemical production of ozone does not occur, the most likely explanation is that this was due to downward mixing of ozone from the previous day's remnant layer. The ozone-loss mechanisms discussed previously occur at the surface (deposition) or within the nocturnal stable layer (titration

by NO). Therefore, at night, it is likely that ozone concentrations will be lower below the inversion than above it. The nocturnal profile for CO can be opposite, as CO can build up beneath the inversion during the night because of local traffic sources [Mao and Talbot, 2004b]. Therefore downward vertical mixing at night can bring air with relatively higher ozone and lower CO down to the surface.

[29] Coincident with the rise in ozone and drop in CO, the meteorological data at TF indicated an abrupt wind shift, an increase in wind speed, and a slight increase in temperature. A nocturnal increase in temperature is a sign of turbulent

mixing, and both *Nappo* [1991] and *Reitbuch et al.* [2000] saw nocturnal increases in both temperature and ozone which they attributed to turbulent mixing. The changes in wind indicated a disturbance that could initiate vertical mixing, as shown by *Darby et al.* [2002]. Doppler lidar measurements from Rye Harbor (Figure 1), discussed in more detail in a later section, indicated the formation of a nocturnal low-level jet by 2200 LST on 4 August (Figure 7b, first panel) and OPAL data indicated high concentrations of elevated ozone at about the same time (Figure 6). There could have been enhanced mixing associated with the low-level jet initiating downward mixing of air with higher ozone and lower CO values, relative to surface measurements [*Banta et al.*, 2006]. Although the Doppler lidar was ~ 20 km southeast of TF and the NOAA research vessel *Ronald H. Brown* was ~ 53 km southeast of TF (see asterisk in Figure 9b indicating the R/V *Ronald H. Brown*'s position), with neither necessarily representing the conditions at TF, the measurements do give a general idea of the possibilities for the regional situation. This nocturnal rise in ozone at TF is important because it kept the surface ozone concentrations from declining to low levels during the night, resulting in enhanced morning ozone levels at TF.

3.3. Mixing Heights

[30] It has been hypothesized that a difference in mixing heights between TF and ADI may in part account for the ozone differences between the two sites [*Mao and Talbot*, 2004a]. Mixing heights are strongly related to surface properties. Since ADI is surrounded by water and TF is on land, the heights of the respective mixed layers are driven by different surface temperatures which have different responses to the diurnal heating cycle due to differences in the specific heat of land and water.

[31] Wind profiler signal-to-noise ratios (SNR) are commonly used to establish mixing heights for sites over land [*Angevine et al.*, 1994; *White et al.*, 1999]. For sites over water, the situation is complicated by the formation of a shallow, often less than 300 m, stable boundary layer over the cooler water. The continental boundary layer, with heights well over 1 km, will be advected over the water, and over the shallow stable boundary layer, when the winds are offshore. The SNR from a profiler over water will then indicate the height of this advected continental boundary layer, but a radar wind profiler may not detect the height of the shallow stable layer at the water's surface, as the marine boundary layer often forms below the profiler's minimum

range. Fortunately, radiosondes launched from the R/V *Ronald H. Brown* during NEAQS-2002 indicate the characteristics of this marine boundary layer near ADI.

[32] Figure 8 shows radar wind profiler SNR for two sites, ADI and the inland station of Concord, NH (CCD,

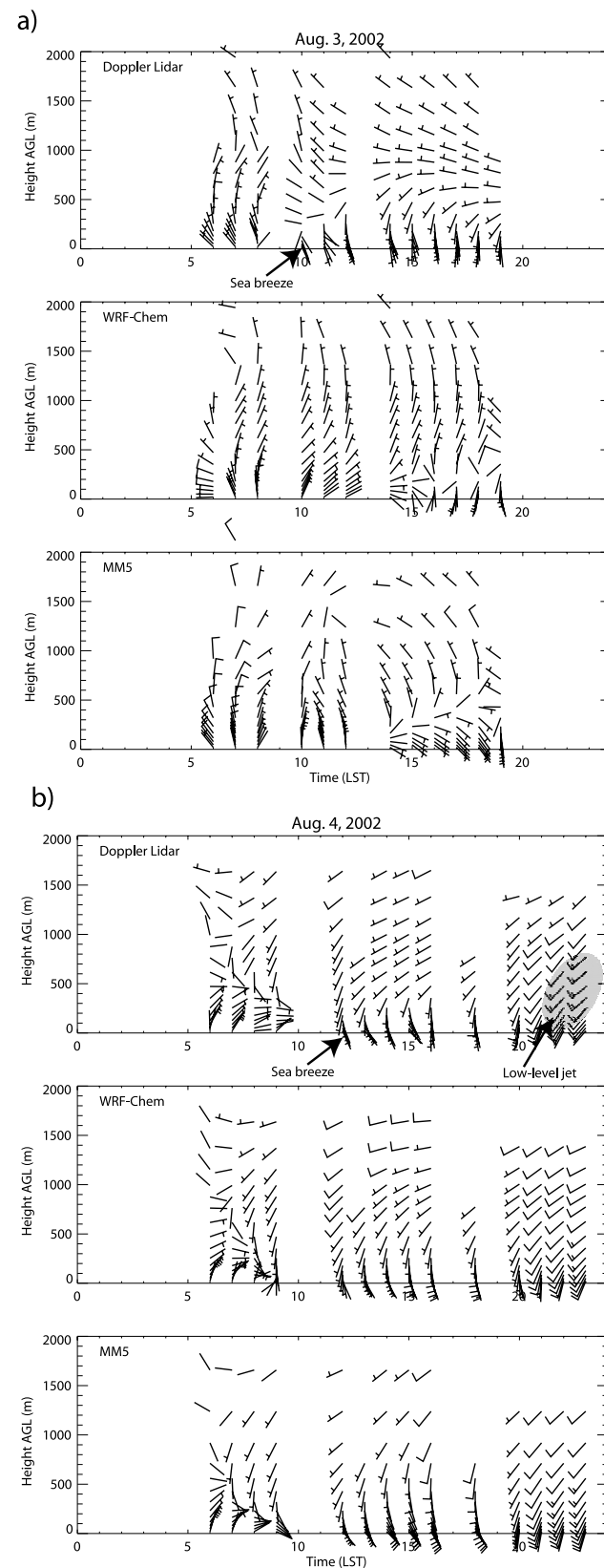


Figure 7. Modeled and observed wind profiles. The miniMOPA Doppler lidar was located at Rye Harbor, ~ 10 km from ADI (Figure 1). The WRF/Chem and MM5/Chem profiles were extracted from the respective grid point closest to ADI. (a) 3 August 2002. (b) 4 August 2002. (c) 5 August 2002. (d) 6 August 2002. For each date, the first panel shows Doppler lidar, the second panel shows WRF/Chem profiles, and the third panel shows MM5/Chem profiles. Wind barbs point into the wind. Half-barbs represent 5 m s^{-1} wind and full barbs represent 10 m s^{-1} wind. Doppler lidar measurements were binned to match the model heights and then averaged. Light gray shading in Figure 7b indicates low-level jet.

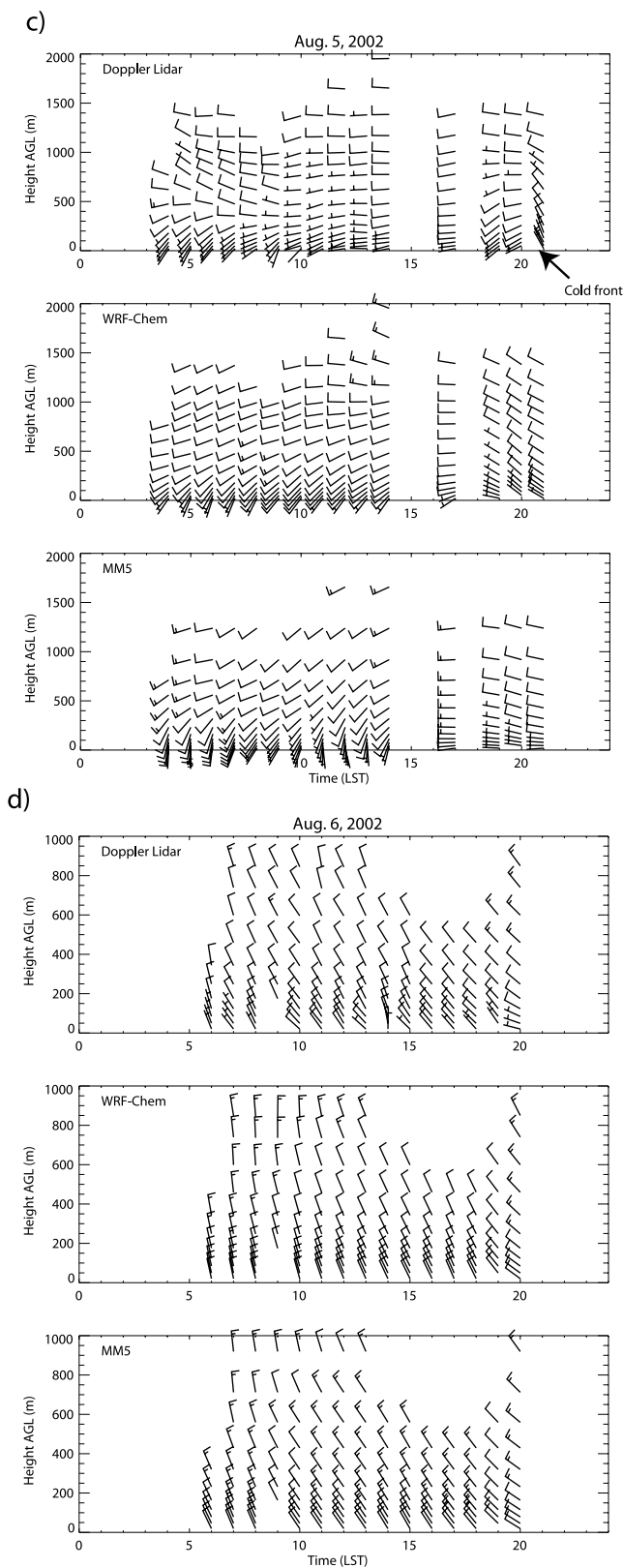


Figure 7. (continued)

Figure 1) for our four days of interest. (Unfortunately, data were missing for the afternoon of 4 August at ADI.) Mixing heights can be inferred from these plots. The enhanced signal above ~ 57 dB (yellow and red pixels) indicates the

height of the continental boundary layer, or mixing layer, as advected over ADI (left side) and at CCD (right side). Doppler lidar profiles (Figure 7) indicated an offshore flow component throughout most of the mixed layer for these afternoons, indicating that the continental boundary layer was, in fact, advected offshore.

[33] On 3 August, after 1100 LST, the afternoon continental boundary layer heights were almost the same at the two sites (~ 2.4 km AGL), implying a similar continental boundary layer height over TF. Potential temperature θ , calculated from R/V *Ronald H. Brown* sondes, Figure 9a, also indicated an afternoon inversion at ~ 2.4 km AGL. Below 500 m, the θ profiles indicated a stable marine boundary layer. These profiles present evidence of the continental boundary layer overriding the marine stable layer.

[34] Afternoon SNR values (proportional to the refractive index structure function parameter of turbulence theory) beneath the continental boundary layer over ADI were much smaller than at CCD. This indicated that while the continental boundary layer was advected over the water, the strong mixing that occurred over the land, as suggested by the higher SNR values at CCD, was suppressed by the cooler surface of the marine environment and the marine inversion. θ profiles on 4 August 2002 (Figure 9b) indicated a similar structure to 3 August, but with an even more well-defined inversion at the surface and aloft in all profiles.

[35] Enhanced mixing at CCD on 5 August, as indicated by higher SNR values throughout the afternoon boundary layer, was readily apparent. Compared to 3 and 4 August 2002, the R/V *Ronald H. Brown* θ profiles for 5 August (Figure 9c) showed a deeper layer of mixing in the afternoon, a weaker inversion aloft, and no marine inversion in the 1459 and 1800 LST profiles.

[36] On the afternoon of 6 August the top of the continental boundary layer, as indicated by profiler SNR data, was distinct and at similar heights at both sites, implying a similar height at TF (~ 2.1 km AGL, Figure 8d). The R/V *Ronald H. Brown* θ profiles showed a weak inversion aloft, more elevated than on the previous days. Again, enhanced mixing within the boundary layer, relative to ADI, was seen at CCD.

[37] It appears from these plots that a distinct difference in the intensity of the vertical mixing existed between the two sites due to the stable marine boundary layer, and it is the differences in the mixing, not necessarily the differences in the boundary layer heights, that affects the ozone differences between the two sites. Enhanced vertical mixing over the land is likely to reduce ozone concentrations near the surface, whereas the shallow marine boundary layer will trap ozone concentrations in a shallow layer with reduced mixing, increasing surface ozone concentrations at ADI. Analysis of the R/V *Ronald H. Brown* θ profiles are consistent with Angevine et al. [2004] who hypothesized that as higher ozone values are advected toward ADI, over the water, some will remain in a shallow layer close to the surface because of suppressed mixing. This does not eliminate the possibility of ozone transport above the marine boundary layer, however, as evidenced by OPAL data (Figure 6).

4. Radar Wind Profiler Back Trajectories

[38] Back trajectories were calculated from a network of 915-MHz wind profilers that were deployed by NOAA and

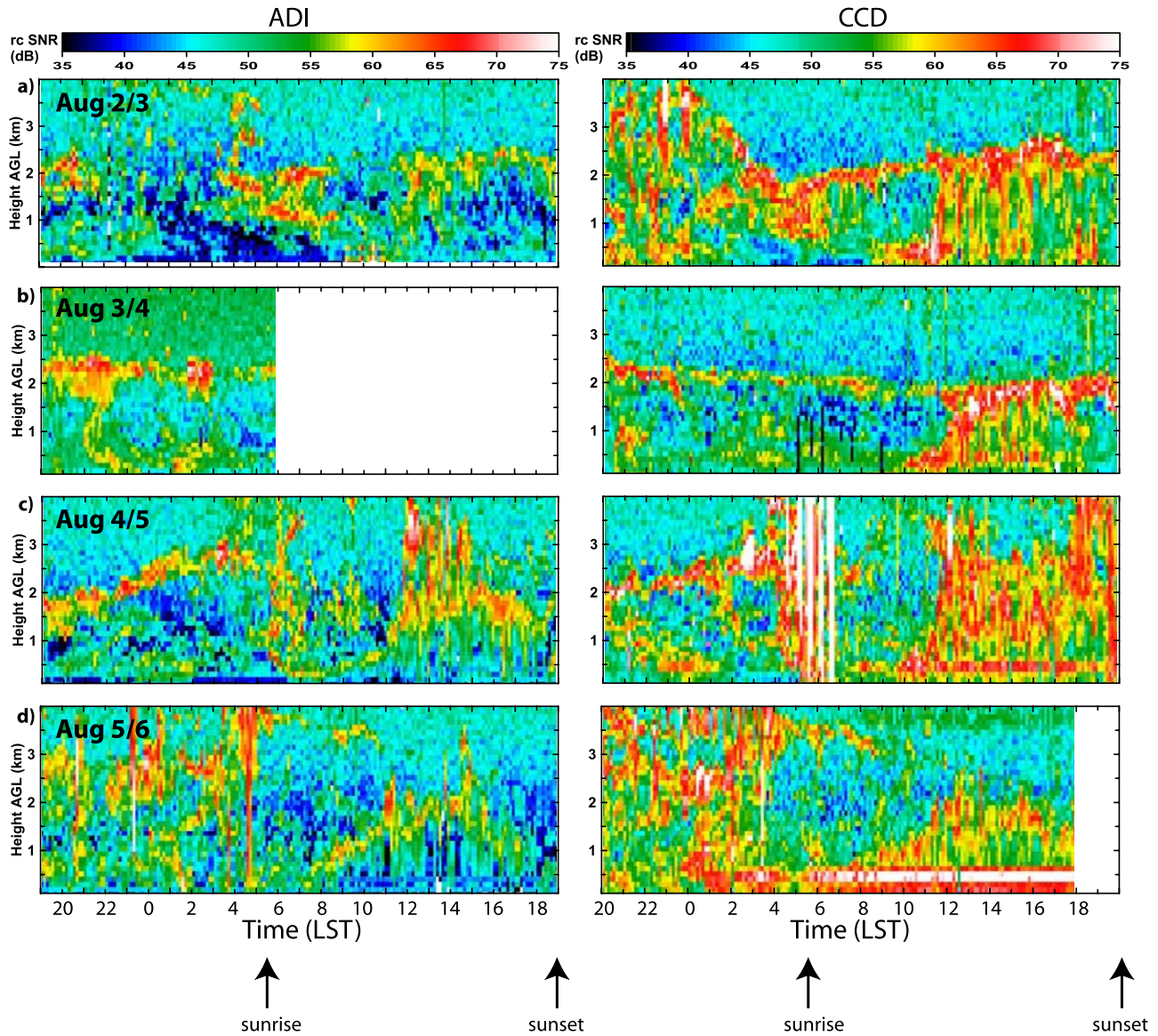


Figure 8. Profiler signal-to-noise ratio (SNR) versus height from the Concord (CCD) and Appledore Island (ADI) profilers. The color scale indicates the intensity of the SNR (dB), from which we can estimate the top of the mixed layer and the vigor of the mixing. (a) 3 August 2002. (b) 4 August 2002. (c) 5 August 2002. (d) 6 August 2002.

other agencies for the NEAQs-2002 field program. These trajectories were calculated for three levels: wind profiler gates below 250 m ASL (the typical lowest gate is ~ 150 m AGL, with 60 m gate spacing), 250–500 m ASL, and 1000 to 1500 m ASL. Additionally, data from ocean buoys and the Coastal Marine Automated Network (C-MAN) stations in the Gulf of Maine were used to calculate trajectories at the surface. Caveats when using back trajectories include (1) small-scale, subhourly wind features will not necessarily be represented in these back trajectories because we used hourly averages of the winds and (2) vertical motion is not accounted for in these trajectories. The back trajectories were computed from wind data from all profilers in the network using an inverse square distance weighted average. See *White et al.* [2006] for more detail on the back trajectories.

[39] We assess 24-hour back trajectories that end at ADI and TF. Since the surface trajectories were derived from

buoy data, they were adequate for assessing low-level transport to ADI, but not to TF. Thus we show surface trajectories that end at ADI, and trajectories based on profiler data ≤ 250 m ASL and between 250 and 500 m ASL for the TF endpoint. The trajectories calculated from data ≤ 250 m ASL ending at TF represent the lowest level of transport to TF that we can use with confidence. The trajectories between 250 and 500 m ASL represent transport at that level for both sites, as there was not much, if any, variation between back trajectories at this height for each endpoint.

[40] Figure 10a shows the back trajectories for 3 August 2002. 1400 LST represents conditions 2 hours before the sea breeze onset at TF. Back trajectories indicated direct transport to ADI from Boston at the surface (black dots), whereas low-level transport to TF was not from Boston. The turn in the ADI surface back trajectory 3 hours before the last

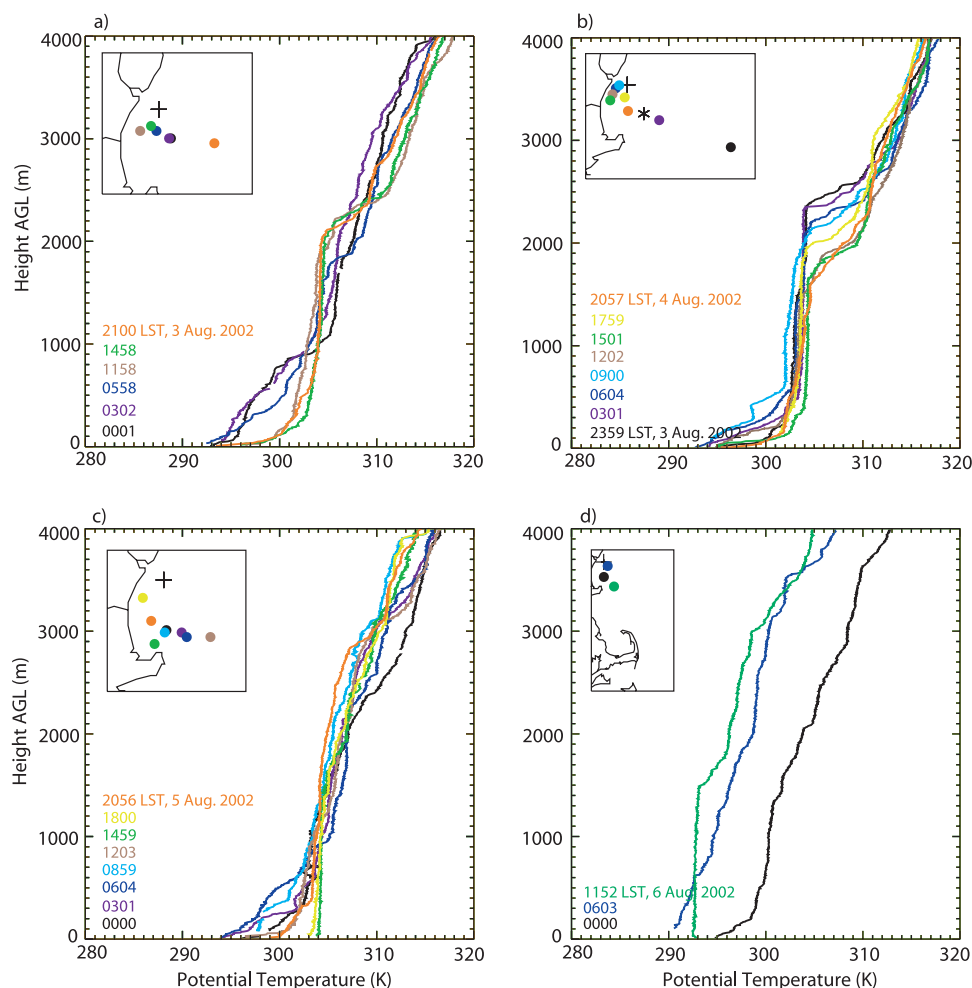


Figure 9. Potential temperature (K) profiles calculated from radiosondes launched from the R/V *Ronald H. Brown*. Each radiosonde profile is color-coded with the dot on the map indicating the location of the R/V *Ronald H. Brown* at launch, the time of which is indicated in the matching color. Radiosonde profiles are grouped by date (LST). (a) 3 August 2002. (b) 4 August 2002. Asterisk indicates position of R/V *Ronald H. Brown* during the time of the nighttime ozone increase discussed in section 3. (c) 5 August 2002. (d) 6 August 2002.

dot indicates the sea breeze onset. Back trajectories were consistent with the surface data (Figures 3, 4a, and 5a), and the overall pattern of these back trajectories indicated that ADI and TF were indeed in different transport pathways.

[41] The back trajectories depict the complete change in character of the transport between 3 and 4 August 2002. On 4 August, back trajectories (Figure 10b) indicated that instead of steady, regional transport from Boston to ADI, as occurred on 3 August, the transport to ADI was only from open water, at the surface, and TF back trajectories indicated a recirculation pattern throughout the 24 hours. Analysis of the surface pressure field (not shown) indicated a mesoscale low centered off the shore of Cape Ann, MA, supporting the recirculation pattern. The analysis in section 3 indicated that each site was in a different transport pathway, yet the back trajectories and surface analysis show a more complicated picture for this day. The day of 4 August was a high ozone day at ADI. Possible reasons for this, based on the back trajectories, include (1) accumulated ozone from the day before was transported to ADI from the open water (since there is much less deposition

over the water than over land) and then landward to TF with the sea breeze; (2) enhanced ozone production over water due to the accumulation of precursors from the day and night before, and then transport to ADI and TF; or (3) enhanced transport toward ADI from Boston, above the surface but within the strong shallow inversion layer, and then downward mixing of ozone. The downward mixing is difficult to prove with the available data set. However, given the high ozone at ADI on this day, the transport from near BOS aloft (as shown in the back trajectories and implied in the Doppler lidar wind profiles), the lack of transport from BOS at the surface, and the elevated ozone concentrations approaching 100 ppbv in the region as measured by OPAL (Figure 6), the possibility of downward mixing on this day cannot be discounted.

[42] In Figure 10c the effects of the coastal trough are seen for 5 August, with a more southerly component to the trajectories leading to ADI, and a more westerly component for the TF trajectories, consistent with the analysis shown earlier. Although much of the time the surface trajectories were over land, where no inland measurements were used to

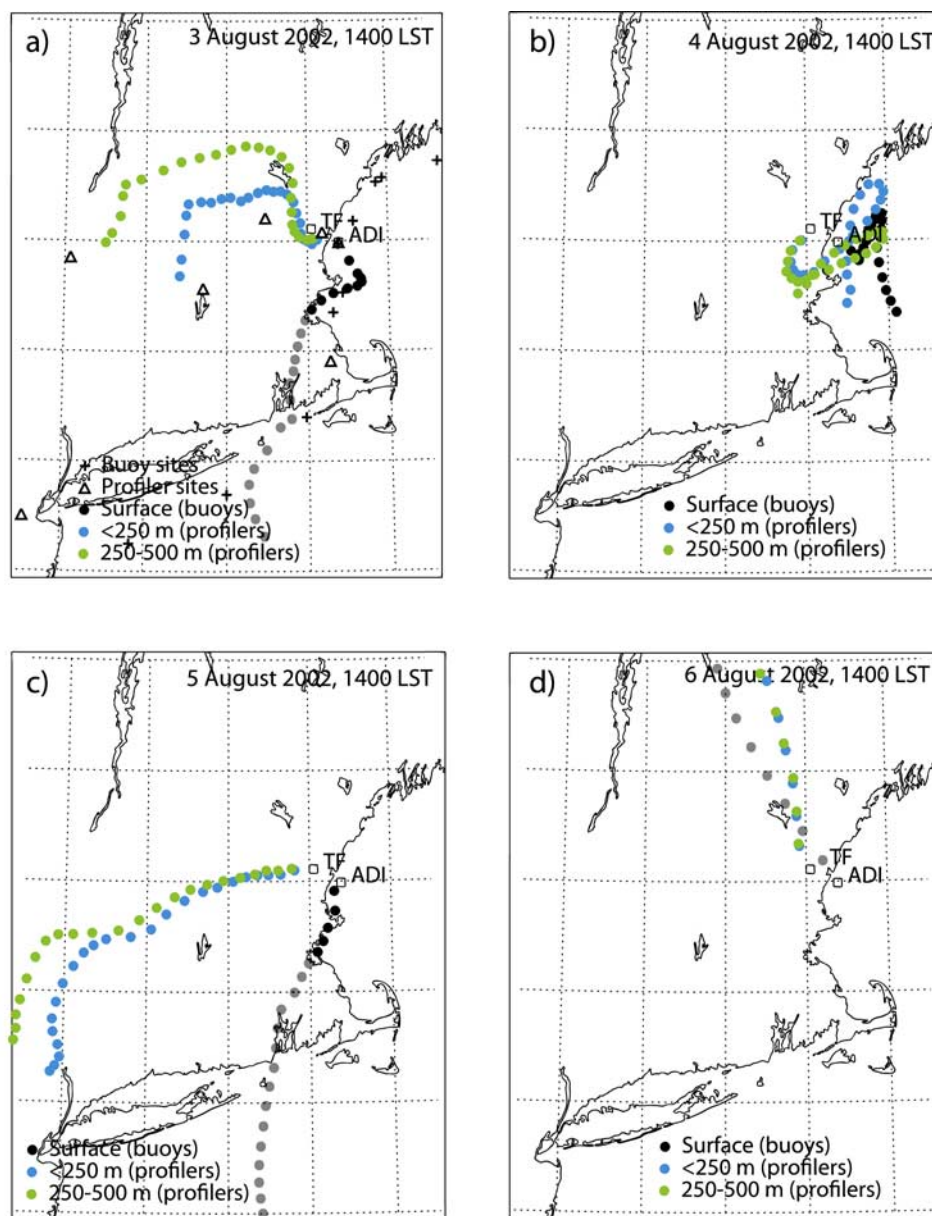


Figure 10. The 24-hour back trajectories calculated from the 915-MHz radar wind profiler network deployed for the NEAQs-2002 field campaign and from wind data acquired from permanent buoys and coastal stations. Triangles represent locations of wind profiles used to calculate the back trajectories above the surface. Crosses indicate positions of the buoys closest to ADI used in the surface back trajectory calculations. Wind profiles from a profiler deployed on the R/V *Ronald H. Brown* were also incorporated in the back trajectories. The dots represent the position of the air parcel 1 to 24 hours before the end time of the trajectory, which is 1400 LST. Black and gray dots represent trajectories calculated at the surface using buoy and coastal station data, ending at ADI. The gray dots indicate the portion of the trajectory that should be interpreted with caution since no land stations were used in the calculations. Blue dots are the back trajectories calculated from wind profiler gates <250 m ASL, with TF as the end point. Green dots represent trajectories calculated from wind profiler gates between 250 m and 500 m ASL, end point also at TF. Dots that fell outside of the map boundary were deleted. (a) 3 August 2002. (b) 4 August 2002. (c) 5 August 2002. (d) 6 August 2002.

calculate them, they do imply a steady south to southwest flow throughout the day. At 1400 LST, ADI was experiencing direct transport from Boston, with coincident ozone concentrations >80 ppbv. The sudden drop in ozone at ADI between 1400 and 1500 LST (Figure 3a) coincided with a shift in the surface trajectories to the southwest, bypassing

downtown Boston. Back trajectories in Figure 10d, for 6 August 2002, indicated northwesterly transport behind the front, as expected.

[43] We looked at back trajectories from days with similar ozone maxima above 80 ppbv at both the ADI and TF sites (the “blue star” days seen in Figure 2c) to help determine

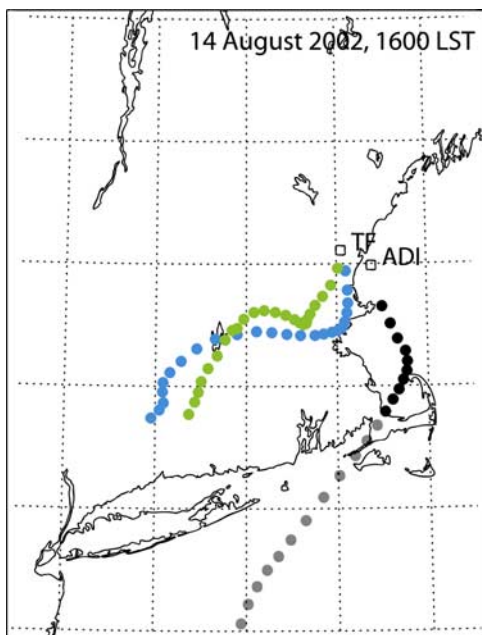


Figure 11. As in Figure 10 except for 1600 LST, 14 August 2002.

what was different between the days with similar ozone maxima and those with a large difference between the maxima (“red star” days in Figure 2c). Figure 11 shows results for 14 August 2002, a sea breeze day with the highest ozone of the summer and nearly the same ozone maximum at both ADI and TF. TF had direct transport from Boston indicated in the trajectories ending at 1600 LST, which enhanced the ozone concentrations at TF, relative to ADI, which, curiously, did not have direct transport from Boston at the surface, again showing evidence for ozone transport above the surface, and then downward mixing at the island site. Cluster analysis of back trajectories from all of July and August (described by *White et al.* [2007]) indicated that this direct transport of ozone from BOS to TF was more likely to occur on days when the ozone maxima were nearly the same at ADI and TF.

[44] Overall, the back trajectories calculated from the radar wind profilers summarized the characteristics of the transport for any given day, and showed the differences in transport due to mesoscale meteorology that can occur from day-to-day. In particular, the back trajectory analyses indicated if TF and ADI were indeed in different transport pathways (e.g., 3 and 5 August), a similar pathway (e.g., 6 August), or if the situation was more complex (e.g., 4 August).

5. Model Performance

[45] Numerical prediction models are being developed for the purpose of predicting surface pollutant concentrations at specific locations. Key aspects of this problem are to be able to differentiate between nearby sites with different meteorological effects, and between days with different meteorological conditions. In the preceding sections we presented analyses of case studies which highlighted these differences. In this section we compare these case study data with output from two chemistry-transport models.

[46] From an air quality point of view, the weather pattern evolved substantially from 3 to 6 August 2002. We have shown that various mesoscale meteorological features caused ADI and TF to be in different transport pathways leading to differences in ozone concentrations. How well did numerical weather prediction (NWP) models do in simulating the chemistry at TF and ADI, and the winds at ADI? With the changes in the meteorological situation, it was hypothesized that the numerical models would increasingly perform better with time over the four-day period, more accurately predicting both winds and chemistry. It was also hypothesized that the models would have a more difficult time correctly predicting the thermally forced sea breeze on 3 and 4 August, as opposed to predicting the large-scale postfrontal northwesterly flow of 6 August.

[47] We looked at the performance of two models, the Weather Research and Forecasting/Chemistry model (WRF/Chem) and the Mesoscale Meteorological Model/Chemistry, version 5 (MM5/Chem) which were previously evaluated using photochemical and aerosol data from the NEAQS-2002 field study [*Grell et al.*, 2005]. Over the summer of 2002, the WRF/Chem model showed statistically better correlations than MM5/Chem in forecasting O₃ and reactive odd nitrogen (NO_y) at all surface sites analyzed. However, both MM5/Chem and WRF/Chem had high biases in CO and NO_y with WRF/Chem having significantly higher biases relative to MM5/Chem. These CO biases were also evident in this case study, as shown in the time series comparisons in Figure 12. Here we focus on modeled and observed relative CO changes with regard to the occurrence

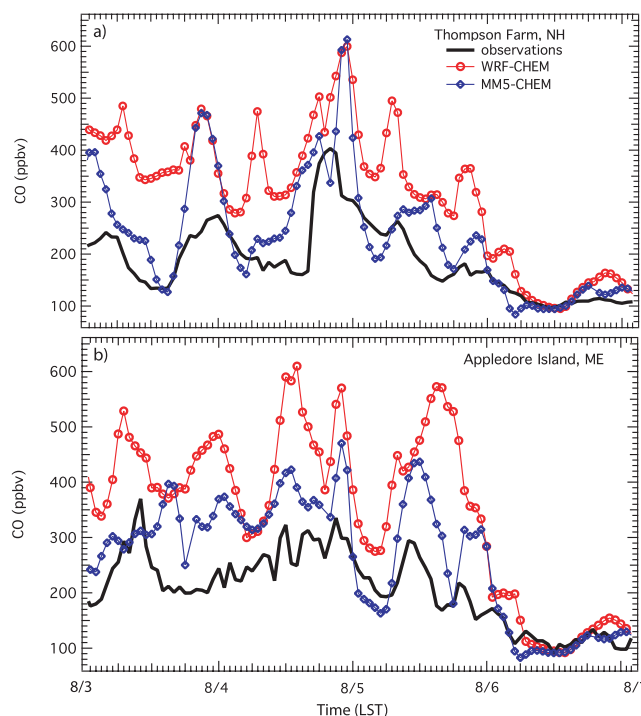


Figure 12. Carbon monoxide (CO) predictions for TF and ADI from the WRF/Chem and MM5/Chem models and hourly averaged CO observations for 0000 LST 3 August 2002 to 0000 LST 7 August 2002. (a) Thompson Farm and (b) Appledore Island.

of sea breezes and a frontal passage. Direct ozone comparisons for this time period were difficult to compare and interpret because of NO titration from anthropogenic NO_y sources, and the fact that model NO/NO_y concentrations were biased high analogous to CO. Model output from the grid points, which had 27-km horizontal spacing, closest to ADI and TF were chosen for comparisons to CO measurements at those locations and modeled wind output from the closest grid point to ADI were chosen for comparison with Doppler lidar wind data.

[48] Figure 7 compares modeled wind profiles with Doppler lidar wind profiles. The Doppler lidar data were binned to match the height levels of each model, with the profiles matching the WRF/Chem model shown here. Given the importance of the low-level winds in transporting pollution, it is necessary to have detailed winds in the lowest 200 m above ground level (AGL) to assess model performance. Doppler lidar measurements are able to provide winds close to the ground, and at high resolution. This is an improvement over using 915-MHz radar wind profiler winds for model evaluations, since the lowest usable gate may be at 150 m AGL, or above. Only model times and heights that corresponded to lidar times and heights were plotted, for ease of comparison.

[49] On 3 August the onset of the sea breeze at the lidar site at Rye Harbor was between 0900 and 1000 LST. (Onshore southeasterly flow is indicated in the 1000 LST profile, which is an average of all profiles between 0900 and 1000 LST.) As shown previously, the time of the sea breeze onset at ADI was 1130 LST and at TF was 1600 LST. Both WRF/Chem and MM5/Chem modeled the sea breeze onset at ~1400 LST, which was later compared to the coastal observations, but two hours before the onset at TF. The modeled winds above the sea breeze had too much of a northerly component. On 4 August, however, both models predicted the onset of the sea breeze much earlier, in agreement with the actual onset at the lidar site and ADI, and modeled winds above the sea breeze were also much more realistic on 4 August than on 3 August.

[50] The Appalachian trough day, 5 August, had better wind predictions early in the morning, particularly by WRF/Chem, but both models missed the onset of steady low-level westerly flow at 1200 LST. The WRF/Chem model did capture the switch to northwesterly flow associated with the cold-front passage with fairly good timing. Both models did a good job predicting the postfrontal winds on 6 August, although they overpredicted the wind speeds, especially MM5/Chem.

[51] Several features of the modeled CO time series at TF (Figure 12a) were directly related to modeled winds, while others were related to model peculiarities. An example of the latter are the CO peaks near 0800 LST at TF on 3, 4, and 5 August within the WRF/Chem model. These sharp peaks were analogous to large, spurious peaks in PM_{2.5} aerosol concentrations at suburban and urban locations in the northeast U.S. for this particular WRF/Chem simulation [McKeen *et al.*, 2007]. That study also showed the choice of land surface scheme and planetary boundary layer (PBL) transport parameterization within WRF/Chem has a dominant impact on the diurnal cycle of PM_{2.5}, and hence other relatively inert constituents emitted near the surface. The MM5/Chem results in Figure 12, and more recent versions

and configurations of WRF/Chem, exhibited more efficient vertical PBL transport, leading to reduced regional biases and improved diurnal cycles of species such as CO, NO_y and PM_{2.5}.

[52] Figure 12 shows that both models reproduced the late afternoon/early evening spikes in CO at TF on 3 and 4 August, presumably because of the sea breeze, although local emissions within the stable evening PBL were probably also contributing. Both models also predicted the relative change in this peak from day to day. The timing of the peaks for both models appeared a couple hours early on 3 August. For 4 August the timing was a couple of hours delayed. For 5 August, observations and WRF/Chem displayed an early morning CO spike overlaying a general decrease from roughly 0400 to 1500 LST at TF, while MM5/Chem was generally increasing during this time. This is consistent with the low-level southerly and southeasterly flow within MM5/Chem being too strong (Figure 7c), during the same period. Both models correctly simulated the reduced CO for the post-cold-frontal conditions on 6 August as strong regional forcing dominated the flow originating from cleaner background conditions.

[53] Relative CO concentrations at ADI appeared to be much better correlated for MM5/Chem compared to WRF/Chem. This was expected, since CO levels at ADI were determined by complex interactions between PBL dynamics and the weak meteorological forcing occurring from 3 to 5 August. The previously noted deficiencies in the WRF/Chem PBL transport probably contributed to its relatively large CO fluctuations and reduced correlation relative to MM5/Chem. The fact that both models did a much better job of predicting CO on 6 August, the day with strong synoptic-scale forcing, indicated that the models did a better job at the synoptic scale, but were weaker in simulating the factors responsible for ozone production and transport when meteorological features such as the sea breeze were prominent; that is, the models did not handle the local-scale meteorology as well. There are probably several reasons for the inability of both models to reproduce CO changes attributed to more local-scale phenomena: (1) the horizontal grid spacing of 27 km was insufficient to resolve the meteorological and emission features occurring along a coast line; (2) the models do not resolve ADI which is classified as ocean in both models; (3) resolved scale PBL transport parameterizations uncertainties discussed by Grell *et al.* [2005] and McKeen *et al.* [2007]; and (4) basic meteorological errors within the model formulations that affect winds, temperature gradients, and the timing of events.

6. Summary

[54] Ozone at ADI was, on average, 11.6 ppbv higher than at TF during the months of July and August 2002. The effects of the sea breeze, a lee-side trough, a cold front, and a stationary front were analyzed, using data from a variety of measurement systems, to assess their roles in the ozone differences between the two sites during four days in August. Through the analysis of surface meteorological and chemical data, and transport aloft using radar wind profiler back trajectories, it was found that all of these features had an impact on the horizontal distribution of

pollutants along the New Hampshire sea coast. ADI and TF were in different transport pathways, with westerly winds at TF and southerly component winds for ADI, leading to higher ozone at ADI. This effect could be overcome by the sea breeze, which brought higher ozone concentrations to TF. The cold front “cleaned out” the region by bringing in a new air mass from an area with few air pollution sources.

[55] We analyzed three days when daytime ozone at ADI was at least 20 ppbv higher than at TF, with even larger differences occurring during the night of 3 August. As stated in section 2, possible mechanisms for these higher ozone values suggested by Mao and Talbot [2004a] included (1) more diverse upwind sources for ADI, (2) lower mixing heights over ADI, (3) transport by the sea breeze, and (4) transport of pollutants that had become elevated at the convergence zone of large-scale southwesterly flow and the sea breeze (for instance, at Boston). These mechanisms, determined mainly from model results, were addressed here through extensive analysis of the enhanced measurement deployment during NEAQS-02. Our results, organized by these four hypotheses, are discussed below. Additional reasons for the ozone differences between the two sites, arising from our analysis, follow.

[56] 1. The analysis indicated that the higher daytime ozone values at ADI, relative to TF, coincided with the presence of a surface trough near the coast, which caused TF to have westerly component flow while ADI had southerly component flow, confirming that ADI and TF had different source regions leading to differences in maximum ozone. TF was often in the pathway of relatively “cleaner” westerly flow, whereas the southerly flow at ADI included the accumulation of pollution from many upstream sources.

[57] 2. Radar wind profiler SNR plots and the R/V Ronald H. Brown θ profiles confirmed that not only were there mixing height differences between the sites, but that the mixing over land was more vigorous than over the water. The combination of these two features was most likely partially responsible for higher surface ozone measurements over the water.

[58] 3. As for the sea breeze at ADI, it was not apparent in the cases analyzed here that higher ozone occurred at ADI because of the sea breeze, but rather from longer-range transport. Since the sea breeze did bring ozone-rich air to TF, however, the sea breeze can act to overcome the ozone differences at the sites due to items 1 and 2.

[59] 4. Analysis of two high ozone days (4 and 14 August 2002) suggests that winds aloft played a prominent role during some high ozone events. Back trajectories for these two days (Figures 10b and 11) indicated that afternoon transport from source areas toward ADI occurred above the surface, not at the surface. On this type of day we expect that ozone was mixed downward at ADI, raising surface ozone concentrations; however we were unable to document this with the available data set. We know from lidar ozone profiles (e.g., Figure 6) that significant amounts of ozone can be transported above the surface.

[60] Additional reasons for higher ozone concentrations at ADI include the accelerated loss of ozone at TF after sunset, relative to ADI, which accentuated the ozone differences between the two sites. The enhanced titration and deposition rates at TF at night caused TF to begin the day

with less ozone than ADI, unless there was nocturnal downward mixing of ozone, as seen late on 4 August 2002.

[61] Overall, the presence of a coastal trough appeared to be the most important factor explaining larger ozone differences between the two sites. This feature was not emphasized as a possible mechanism in Mao and Talbot [2004a]. The analysis presented here illustrates the importance of local meteorology. When mesoscale features, such as the coastal trough or a frontal boundary, dominated the meteorology of this region, quite often the winds and ozone at the two sites were different, in contrast to 6 August, when synoptic meteorology dominated, leading to similar winds and ozone at both sites. The results of the model evaluations indicated that local-scale versus synoptic-scale meteorology is also important for modeling. The models had less skill predicting both winds and CO on days when the local-scale meteorology dominated.

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